COMBUSTION CHARACTERISTICS OF HYDROGEN-PROPANE MIXTURES

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ABSTRACT

An experimental study of the combustion and pollution characteristics of a diffusion flame of hydrogen-propane fuel mixtures is described. Flame appearance, visible length, radiative fraction of heat release, the emission indices of NO, NOx, and CO, and the inflame profiles of temperature and composition are presented. Results are compared for the flames of pure hydrogen, and the hydrogen-propane mixtures with 20% and 35% (volume) of propane.

INTRODUCTION

Because of their increased availability, relatively superior burning characteristics, and low pollutant emission potential, gaseous fuels such as compressed natural gas (CNG) and liquid petroleum gas (LPG) have recently received increased attention as potential fuels for transportation applications. A major problem that is hindering the wide-spread application of these fuels is their storage at a reasonable energy density. The Institute of Gas Utilization Technology (IGUT) at the University of Oklahoma is studying various methods, including dissolving natural gas in heavier hydrocarbons as a means of increasing the energy-density [1]. On another front, hydrogen has been advocated and studied for use in internal combustion engines [2]. The high reactivity and flame velocity of hydrogen offer additional benefits, particularly for the so-called lean-burning engines. Because of the highly nonlinear nature, the combustion characteristics of fuel mixtures cannot be predicted from those of the constituent fuels. Hence, a program to study the application of the fuel mixtures in laboratory flames, in engines on dynamometer test stands, and in actual automobile vehicles is in progress at the IGUT. This paper, one of a series of papers [3, 4] on mixed fuel characteristics, deals with the combustion of hydrogen-propane mixtures in a diffusion flame.

EXPERIMENTAL DETAILS

Experiments were conducted in a vertical steel combustion chamber of 76 cm × 76 cm cross section and 163 cm height. The chamber was fitted with rectangular windows of dimensions 20 cm × 20 cm ×145 cm on all of its four side walls. Three of the windows were fitted with Pyrex plate glass and the fourth was fitted with a slotted metal sheet for introducing probes. Air was induced by natural convection into the test chamber through a 20 cm diameter circular opening in the base plate. Three layers of fine-wire-mesh screens were used to provide a uniform flow. The circular fuel burner used in these experiments consisted of a stainless steel tube of 2 mm ID, which injected fuel into an atmosphere of air. The burner projected 14.5 cm above the chamber floor.

Propane and hydrogen were supplied from cylinders, through pressure regulators, rotameters, a mixing chamber, and in-line filters. The two fuels were mixed inside an annular mixing device in which secondary fuel was injected into the stream of primary fuel through a concentrically located injector. The length of the mixing device was sufficiently large (>150 hydraulic dia.) to ensure the homogenous mixture of fuels. For a fixed jet exit Reynolds number, the volume flow rate of the hybrid fuel was calculated for different mixture conditions. The volume flow rates of the primary and secondary fuels were then regulated with calibrated rotameters. The fuels used were of commercial grade with 98% purity.

The instrumentation included a computer-controlled-thermocouple based data acquisition system, a computer-aided precision two-dimensional positioning mechanism, various gas analyzers, a radiometer, a low-energy He-Ne laser for soot concentration, and a high speed camcorder (with strobe and back-illuminating option). For measuring exhaust emissions, a quartz flue gas collector was mounted over the visible flame, and axially aligned with the burner . A sample profile across the collector diameter showed a variation of less than 1.5% in species concentration, and hence, the center point data were treated as the average representative values. Gas samples were collected from combustion products through an uncooled-quartz probe of tip diameter 1 mm and treated to remove particulate and moisture with a series of filters and an ice-chilled moisture trap. The sampling flow rate was adjusted such that the suction and local free stream velocities in the flow-

field were close enough to ensure *quasi-isokinetic* sampling [5]. Chemiluminescence was used to measure the concentration of NO and NO_x. Two non-dispersive infrared (NDIR) analyzers were used to measure the concentration of CO and CO_2.

The species concentrations inside the flames were measured with another uncooled quartz probe. The inside diameter of this sampling probe was increased from 0.5 mm at the probe inlet to 6 mm over a short distance of 20 mm which allowed a sudden expansion of the gas sample, thus freezing its composition. The sample was analyzed using the same analyzers used in the exhaust emission measurements. Neglecting the gradient broadening effect and lack of quenching, the uncertainties in the species concentration measurements are estimated to be less than 9% of the mean value. Temperature profiles in the mixed fuel flame were measured with a Platinum-Platinum 13% Rhodium (Type R) in-house-made L-shaped thermocouple probe with the wire diameter 127µm and bead diameter 280 µm. To measure the temperature in 100% hydrogen flame, a Tungsten-5% Rhenium vs. Tungsten-26% Rhenium (Type C) thermocouple with wire diameter 127 µm and bead diameter 370 µm was used. Since this thermocouple rapidly deteriorates under oxidizing conditions, a coating of high temperature ceramic cement was used on all exposed parts of the thermocouple except the bead. The output was sampled at 1 kHz over a period of 20 seconds, and on-line averaged over 1 second using a computer-controlled high-speed data acquisition system hardware and a PC based data acquisition software. Flame radiation was measured with a wide-angle (150°) highly sensitive pyrheliometer with absorptivity of 0.96.

The visible flame height was determined with a high-speed video camera. Strobe recording technique (1/6 second-interval progression, 1/2000 s.) along with back-light illuminating and DEIS (digital electronic image stabilization system) method were used to visualize the flame image in a dark background. A modified version of the technique proposed by Yagi and Iino [6] was used to measure the soot concentration. A He-Ne laser beam was passed through the flame and due to the presence of soot, the beam intensity was attenuated. The amount of attenuation was measured using a pyro-electric laser power meter placed on the other side of the beam. Table 1 shows the nominal experimental conditions and Table 2 shows the estimated uncertainties in measurements.

RESULTS AND DISCUSSION

Appearance and Flame Length

With the increase of propane concentration in the Hydrogen-Propane hybrid fuel, both flame luminosity and flame lengths increase [Fig. 1]. At Reynolds number =150, pure hydrogen produces a visible flame length of 30.5 mm which increases sharply as the propane concentration increases in the mixture. The Hydrogen-Propane fuel produces a visible flame which is approximately 10% taller than the corresponding Hydrogen-Natural gas flame [7]. The Hydrogen-Propane flames are much more luminous than the Hydrogen-Natural gas flame. Furthermore, the flame shapes are considerably different in both cases. At a Reynolds number = 150 and 80-20% mixture condition, in Hydrogen-Natural gas flame a dull yellow-orange zone appears from the mid-flame region to the far-burner region which is surrounded by a faint blue envelope, whereas in the Hydrogen-Propane flame, the whole far burner region is luminous yellow at that condition. This change in appearance is due to the increased soot formation in the Hydrogen-Propane flame. The chemical structure of propane is favorable to form PAH (polycyclic aromatic hydrocarbons) which is now believed to be the cause of soot inception. At Reynolds numbers of 1000, and 3000 the *soot wings* and the *soot breakthroughs* are more prominent than those in the Hydrogen-Natural gas flames. The trend of flame length increase with higher propane concentration in the fuel mixture is comparable to Roper's correlation [8].

Radiative Heat Loss Fraction

At the Reynolds Number of 150 the radiative heat loss fraction increases from 3.5% for 100% H_2 to 7.2% for 80-20% Hydrogen-Propane flame[Fig. 2]. After that, it does not change appreciably with the increase of propane concentration in the mixture. The increase of radiative heat loss factor is certainly due to the increased radiation from both molecular (CO_2) band and continuous sources (soot particles). However, the asymptotic behavior of flame radiation factor at concentrations more than 20% of propane in the mixture reveals that increased soot formation beyond a certain concentration increases self absorption of radiation between the soot particles [4]. At the Reynolds numbers of 1000 and 3000, this behavior is extremely significant. The flame radiation factor at these Reynolds numbers increases sharply below 20% propane concentration in the mixture. Beyond that concentration, although the flame radiation factor increases, it occurs at a slower rate than that at lower concentrations. At all Reynolds number and mixture conditions, Hydrogen-Propane flames have higher radiation factors than those of Hydrogen-Natural gas flames, as expected.

Emission Indices (EI)

Measurements of emission indices of NO, NO_x, and CO of Hydrogen-Propane flames at different Reynolds Number and mixture conditions are shown in Fig. 3. Only the results at a Reynolds

number =150 are shown here. The results at higher Reynolds numbers (1,000 and 3,000), which exhibit the same trends as those at Re =150 are available in Choudhuri's thesis [7].

NO Emission Index(EI_{NO}): With the increase of propane concentration in the mixture, at all Reynolds numbers, the NO emission index decreases sharply. At Reynolds number =150, the emission index of NO has a value of 0.8 g/kg at the baseline condition (95%-5% Hydrogen-Propane) which decreases to 0.45 g/kg at 80%-20% Hydrogen-Propane mixture. It continues to decrease and then attains a value of 0.28 g/kg for the 65%-35% Hydrogen-Propane mixture. It is notable that the Hydrogen-Propane flame has lower NO emission indices than those of the Hydrogen-Natural gas flame. As mentioned earlier in a high temperature diffusion flame the *Thermal-Zeldovich* mechanism is the primary route of NO formation. Since this mechanism is highly temperature dependent and the Hydrogen-Propane has lower effective flame temperature than the Hydrogen-Natural gas flame these results are expected.

NO_x Emission Index (EI_{NO_x}): The emission index of NO_x follows the same trend as the emission index of NO at all Reynolds numbers. It is also evident that a very small amount of NO_2 is formed in the Hydrogen-Propane hybrid fuel flame. This occurs because an increase of propane concentration in the mixture reduces the concentration of intermediate radicals, OH and O, which are essential to form NO_2. Furthermore, the increased propane concentration increases the radicals like CH and H which ultimately remove NO_2 from the flame. At a Reynolds number =150, the emission index of NO_x has a value of 0.9 g/kg at the baseline condition which decreases to 0.34 g/kg for the 65-35% Hydrogen-Propane mixture. This indicates that the emission index of NO_2 which has a value of 0.1 g/kg decreases up to 60% with the increase of 30% concentration of propane in the mixture.

Carbon monoxide Emission Index (EI_{co}): The carbon monoxide emission index increases with the increase of propane concentration in the mixture at all Reynolds numbers. This is expected, since adding more propane in the mixture means introducing more carbon atoms into the flames which results in an increase of CO. The carbon monoxide emission index (at Reynolds number =150) increases from a value of 0.3 g/kg at the baseline condition to a value of 0.9 g/kg for the 65%-35% Hydrogen-Propane mixture. However, for Hydrogen-Propane flames the carbon monoxide emission index has higher values compared to the Hydrogen-Natural gas flame. This is reasonable since propane has a higher carbon/hydrogen ratio than natural gas.

Volumetric Soot Concentration (*w*)

Volumetric soot concentrations measured at different axial locations and Reynolds numbers are shown in Fig. 4. Similar to the Hydrogen-NG flames, the Hydrogen-Propane flames have also high soot concentrations in the mid-flame region. As mentioned earlier, mid-flame region is the location of soot inception and growth. However, Hydrogen-Propane flames produce more soot than Hydrogen-Natural gas flame. Also, it is evident that soot formation increases sharply for the increase of propane concentration from 20% to 35%. This phenomenon is consistent with the earlier explanation given for the trend of the flame radiation factor. At a Reynolds number =150, 80%-20% Hydrogen flame has a peak soot concentration of 1×10^{-7} g/cc which jumps to 9.5×10^{-7} g/cc with the increase of 15% concentration in mixture. In fact, from the soot concentration point of view, 65-35% Hydrogen-Propane flame behaves almost like pure propane flames. This may be related to the complex soot inception and PAH formation in hydrocarbon flames.

Flame Structure

Temperature Profiles: Radial Temperature profiles at different axial locations for the different Hydrogen-Propane mixtures at a Reynolds number of 150 are presented in Fig. 5. In the near-burner region the dual hump nature of temperature profiles are prominent in all of the conditions. In the near-burner region, the peak temperature occurs at the flame sheet, and it is found to be 1980 K for the 80%-20% Hydrogen-Propane mixture and Reynolds number =150. The peak temperature in the near-burner region is in the range of 1980 K to 2000 K compared to 2250 K in the pure hydrogen flame. The addition of 20% propane in the hydrogen reduces the effective flame temperature by more than 250 K. The effective flame temperature drops again with the increase of an additional 15% propane concentration in the mixture. The 65%-35% Hydrogen-Propane mixture has approximately 100 K lower flame temperature than the 80%-20% Hydrogen-Propane mixture. Since close to the burner, soot concentration is not significant, the drop in the peak temperature in the Hydrogen-Propane flame with the increase of propane concentration in the mixture can be attributed to the banded radiation from molecular source (increased CO_2) and the lower energy input due to the higher propane concentration. Close to the mid-flame zone, both the 80%-20% and the 65%-35% Hydrogen-Propane flames show the dual hump temperature profile. Similar to the near-burner region, the temperature in the mid-flame also decreases with the increase of propane concentration in the mixture, again due to the increase of flame radiation and lower energy input. In the far-burner region, temperature profiles show a single peak close to the burner-axis. However, the peak effective flame temperature does not change appreciably with the increase of propane concentration for the mixture from 20% to 35% propane concentration. This is probably due to the higher amount of soot-oxidation which compensates radiation loss locally at higher propane concentrations in the mixture.

Concentration Profiles:

The full set of concentration profiles of CO_2, CO, NO, NO_x and O_2 are available in Choudhuri, 1997. As expected CO_2 concentration profiles were similar to the temperature profiles and oxygen concentration profiles follow the inverse trend of those of CO_2. Further NO and NOx profiles were similar. Hence, for brevity only CO and NOx profiles are shown here (Figs. 6 and 7).

Carbon monoxide(CO): Close to the burner, the CO concentration profiles show peaks in flame sheets for all three Reynolds number and mixture conditions of Hydrogen-Propane hybrid fuel. This reveals that close to the burner, reactions are mostly confined to the fuel-oxidizer interface. Further downstream, the CO concentration profiles show an axial peak which indicate that CO forming at the stochiometric contour at the flame edges start accumulating at the burner axis. At all conditions, in the far-burner region CO has lower concentration compared to mid-flame region. This is understandable since CO is rapidly oxidized between the mid-flame to far-burner region. As expected, Hydrogen-Propane flames have higher CO concentration compared to Hydrogen-Natural gas flames due to the higher carbon input rate.

Nitrogen Oxides (NO$_x$): As mentioned earlier, the Thermal-Zeldovich route is the dominant NO formation mechanism when flame temperatures are more than 1800 K. Since thermal NO is strongly temperature dependent, usually the NO temperature profiles follow the temperature profiles. Hydrogen-Propane flames for 80%-20% and 65%-35% mixtures have the peak flame temperature equal to/more than 1800 K. Hence, it can be expected that only thermal NO formation is active, and hence the concentration profiles follow the temperature profiles. Furthermore, Hydrogen-Propane flames have lower effective flame temperature which results in lower NO concentration than that of Hydrogen-Natural gas flames. The above mentioned trends are significant for all three Reynolds numbers and mixture conditions. Also, it is found that the Hydrogen-Propane mixture has a lower NO_2 concentration than that of hydrogen-natural gas flames. This is because, due to the high concentration of radicals CH, H, NO_2 removal rate is faster in hydrogen-propane flames compare to hydrogen-natural gas flames.

CONCLUSIONS

Flame luminosity and visible length of a diffusion flame of hydrogen-propane mixtures increase with the increase of propane concentration. The radiation fraction of heat release increases initially with the propane concentration and levels off above 30% (by volume). Soot production increases continuously with propane content of the fuel. The emission indices of NO and NOx decrease and the emission index of CO increases with the increase of propane fraction. The peak temperature in the flame decreases monotonically with the increase of propane content.

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Table 1. Nominal Experimental Conditions

Fuel:	Propane (94%+ methane) (0-35% vol.)
	Hydrogen (98%+) (100-65% vol.)
Jet diameter:	2 mm
Jet exit velocity	2.5-157.7 m/s
Jet exit Reynolds number	150-3000
Jet exit Froude number	$420-1.2 \times 10^6$
Ambient Temperature	295 K
Ambient Pressure	104 kPa

Table 2. Estimated Uncertainties*

Measurements	% of Mean Value
Flame Height	15
Emission Index	1.7
Radiative Heat Loss	2
Concentration of NO	7.9
Concentration of NO_x	8.2
Concentration of CO_2	8.6
Concentration of CO	8.8
Concentration of O_2	4
Temperature	1.4
Soot Concentration	6.4

Based on Student's t-test at 95% confidence
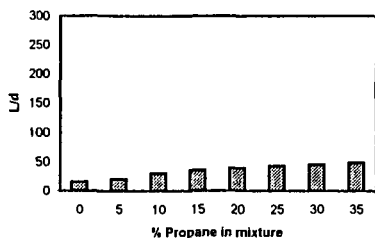
Fig 1: Effect of Propane Concentration (vol.%) on Flame Length (Re=150)
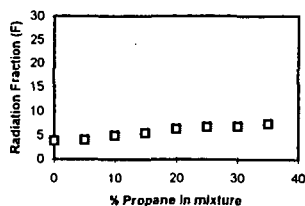
Fig. 2: Effect of Propane Concentration (vol.%) on Radiative Fraction (Re=150)
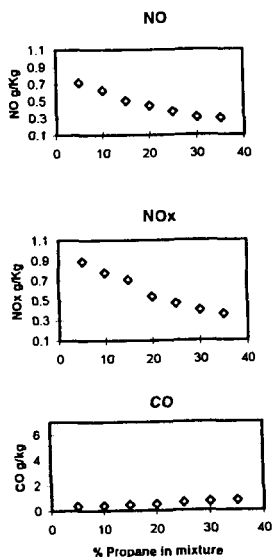

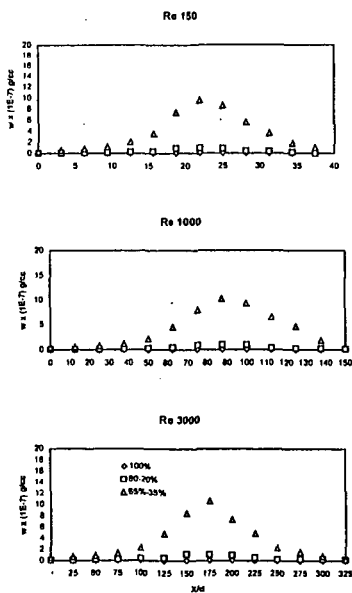Fig. 3 Effect of Propane Concentration (vol.%) on Emission Indices (Re=150)

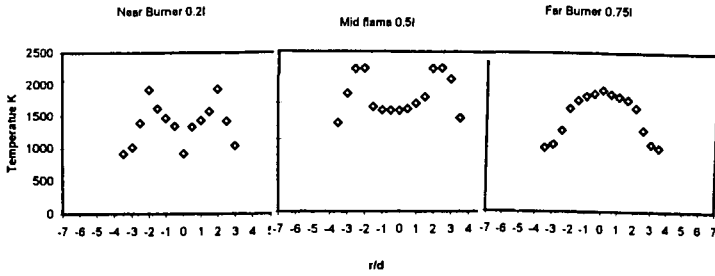Fig. 4: Effect of Propane Concentration (vol.%) on Volumetric Soot Concentration

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Fig. 5: Temperature Profiles in the Near-Burner (x/L=0.33), Mid-flame (x/L=0.5) and Far burner (x/L=0.67) Regions of the Flames (Re=150)
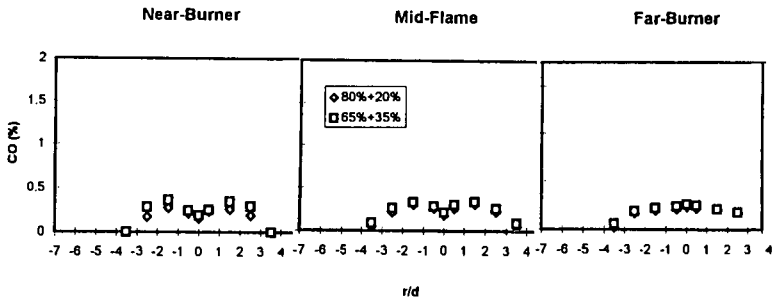

Fig. 6: Concentration Profiles of CO in the Near-Burner (x/L=0.33), Mid-flame (x/L=0.5) and Far burner (x/L=0.67) Regions of the Flames (Re=150)
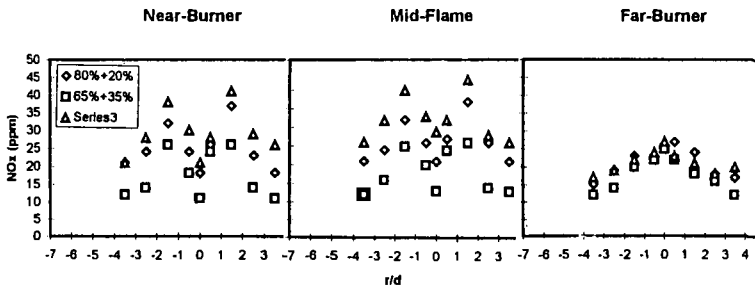

Fig. 7: Concentration Profiles of NOx in the Near-Burner (x/L=0.33), Mid-flame (x/L=0.5) and Far burner (x/L=0.67) Regions of the Flames (Re=150)